

Sulfate-induced cooling in the southeastern U.S.: An observational assessment

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Abstract. The effect of sulfates on cloud microphysical and optical properties have been studied using data from a rural mountaintop location in the Mt. Mitchell State Park, North Carolina. Although the amended Clean Air Act has limited the sulfur emissions beginning 1990, we found the sulfate concentrations greater during 1993-97 compared to that during 1986-89. Cloud albedo inferred from satellite data and calculated from surface observations does not indicate a monotonic increase with increasing sulfates. The direct and indirect (cloud-mediated) sulfate forcing are assessed to be -4.8 and -4 W m⁻². These values exceed current model predictions as evaluated by other investigators. Surface temperature records of the region during 1949-94 indicate a cooling trend tacitly supporting our assessment.

Introduction

It is widely recognized that anthropogenic aerosols perturb the atmospheric radiation field through direct and indirect interactions with solar radiation, thereby influencing climate. Through the direct effect, aerosols can scatter and absorb solar radiation in cloud-free air [Charlson *et al.*, 1992]. In terms of the indirect effect, an increase in aerosol concentration (N_a) composed of soluble substances that act as cloud condensation nuclei (CCN) increase cloud droplet number (N), thereby reducing droplet size, assuming the liquid water content (LWC) stays the same. This enhances cloud albedo [Twomey, 1977] and can suppress drizzle production and increase cloud cover and lifetime [Albrecht, 1989]. Quantification of the indirect forcing is fraught with large uncertainty inherent in the relationship between aerosol number distribution and anthropogenic pollution [Charlson *et al.*, 1992, Penner *et al.*, 1994]. Previous studies estimated indirect forcing by relating sulfate mass to N_a /CCN/N through empirical observations and coupled chemistry-climate models [Jones *et al.*, 1994, Boucher and Lohmann, 1995]. Greater negative forcing is predicted for eastern U.S., southeastern Europe and eastern China [Kiehl and Briegleb, 1992] where sulfate emissions are larger. Empirical evidence of sulfate forcing on surface-air temperatures and optical depth has been previously reported [Saxena and Yu, 1998, Saxena *et al.*, 1997] for the southeastern U.S. Given the regional variability in aerosol properties, long-term regional measurements of aerosol properties in several parts of the globe will be

necessary to reduce the uncertainty in developing appropriate empirical relationships. To investigate the climatic impact of anthropogenic aerosols we have assessed in this paper the direct and indirect effects of aerosols (primarily sulfates and to a lesser extent black carbon (BC)) using surface measurements, modeling results and remotely sensed data. Empirical relations between sulfate, CCN, N and cloud droplet effective radii (R_{eff}) were used to arrive at the relationship between cloud albedo and anthropogenic pollutants. Direct forcing was estimated from a column radiative forcing model (CRM). Our regionally representative observational site intercepts relatively clean marine and highly polluted industrial air masses and being at a remote location, local influences are minimal. This provides unusual and unique opportunities to observe the impact of air mass content on cloud properties. The assessment presented below is not being claimed as final but as the best available to date based on ten years of observations in the southeastern U.S.

Methodology

The field site is located on Mt. Gibbs (2004 m MSL, 35°44' N and 82°17' W), in Mt. Mitchell State Park in North Carolina shown in the upper panel in Fig. 1. The site is at the highest peak in eastern North America, frequently stays in the free troposphere and at least seven out of ten days are impacted by clouds during summer. The types of clouds commonly encountered are those formed due to orographic lifting and those caused by frontal activity that is orographically enhanced. Continuous measurements of meteorological variables were obtained by instruments located atop a 17.1-m walk-up tower, above the forest canopy. A passive string-type cloud water collector mounted on a rotatable carriage atop the tower was used to estimate LWC as in Saxena *et al.* [1989]. A forward scattering spectrometer probe (FSSP) was used to estimate cloud drop-size distribution. The CCN activation spectra were measured by a Horizontal Thermal Gradient Cloud Condensation Nucleus Spectrometer [Fukuta and Saxena, 1979a, b]. The cloud water pH was measured and cloud water ionic species were analyzed. The BC concentrations were measured with a two-channel aethalometer. The air mass sources were analyzed from their back trajectories obtained from the Hybrid Single Particle Lagrangian Integrated Trajectory Model (HY-SPLIT) [Draxler, 1987] and are classified as marine (M), continental (C) and polluted continental (PC) based on the SO_x and NO_x emission inventory produced by the U.S. Environmental Protection Agency, as shown in the lower panel in Fig. 1 [Ulman and Saxena, 1997]. The classification is consistent with high values of BC of 216.6 ± 47.8 ng m⁻³ measured in PC air masses and low values of 65.6 ± 23.5 ng m⁻³ for the M sector [Bahrmann and Saxena, 1998].

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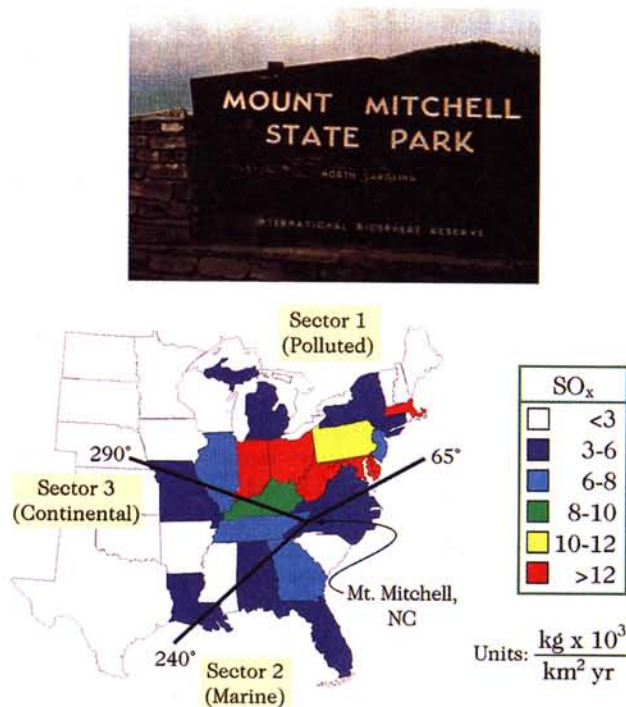


Figure 1. The upper panel indicates the designation of the Mt. Mitchell site as an International Biosphere Reserve by the United Nations. The lower panel shows the 1991 U.S. Environmental Protection Agency emissions inventory of anthropogenic sulfur oxides (SO_x) for the eastern US.

Direct forcing

The direct forcing was estimated by coupling a Mie code to the CRM of the Climate Community Model (CCM III). The sulfate mass in $\mu\text{g m}^{-3}$, estimated from the product of measured cloud water sulfate and LWC, was used in the Mie code to calculate aerosol optical properties. The refractive index for an aerosol composed of 75% H₂SO₄ and 25% water was used, with a density of 1.7 g m^{-3} . A log-normal aerosol size distribution with a mean particle radius of $0.05 \mu\text{m}$ and a geometric standard deviation of 2.1 was used. The aerosol optical properties and the mass mixing ratios were used in the shortwave radiative transfer portion of the CRM to calculate forcing obtained as the difference in the total solar radiation absorbed when the model is run once with and without any aerosol mass loading [Briegleb, 1992; Omar et al., 1999].

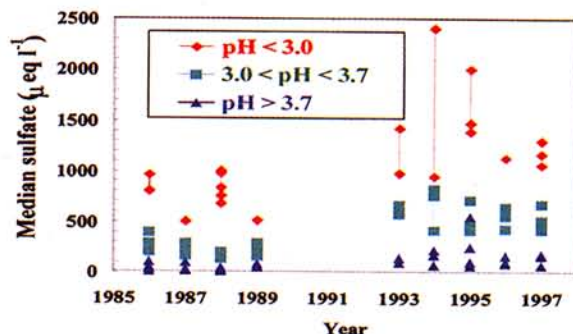


Figure 2. The trend in median values of sulfate for May-August. The data are categorized in terms of cloud water pH. pH < 3.0 represents polluted continental clouds, $3.0 < \text{pH} < 3.7$ continental clouds and pH > 3.7 marine clouds.

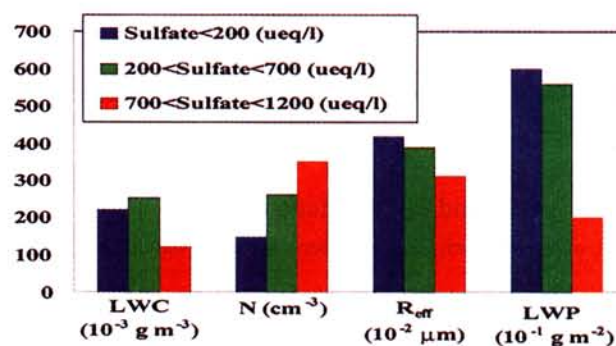


Figure 3a. Average values of cloud liquid water content (LWC), cloud droplet number concentration (N), cloud droplet effective radii (R_{eff}) and cloud liquid water path (LWP) for three categories: sulfate < $200 \mu\text{eq l}^{-1}$ (seven cases), $200 < \text{sulfate} < 700 \mu\text{eq l}^{-1}$ (ten cases) and sulfate > $700 \mu\text{eq l}^{-1}$ (three cases).

During May-August of 1993-96, the direct forcing was calculated for 55 daytime cases. An overall average of -4.8 W m^{-2} was obtained with values of -1.3 , -3.6 and -10 W m^{-2} for M, C and PC air masses, respectively. Since sulfate and BC concentrations are well correlated (suggesting the same source region), the forcing for BC internally mixed with sulfates was calculated for ten cases with coincident sulfate and BC with values ranging from -5.3 to $+0.6 \text{ W m}^{-2}$ for BC-sulfate mass ratios between 0.7 to 10%. The only occurrence of positive forcing was for 10% BC-sulfate mass ratio. The direct sulfate forcing (mass between $1 - 8 \mu\text{g m}^{-3}$) is from -1.1 to -7.2 W m^{-2} , whereas that for BC (mass between $0 - 0.2 \mu\text{g m}^{-3}$) is from $+0.08$ to $+2.1 \text{ W m}^{-2}$. Thus, an internally mixed BC-sulfate aerosol reduces the sulfate forcing by 1.1 W m^{-2} . Modeling studies by Ch_{lek} et al. [1995] indicate that regional effects are important for BC-sulfate ratios of the order of 5% or more. However, despite the reduction in forcing with the addition of BC, the values obtained for sulfate forcing (min -0.07 W m^{-2} to a max -26 W m^{-2}) are much higher than model predictions of -2 W m^{-2} [Boucher and Anderson, 1995] for the southeastern U.S. The above forcing estimates are uncertain by a factor of two due to various assumptions used in the calculation of aerosol optical properties and size distribution.

Indirect forcing

Since the incorporation of pollutants into clouds can affect drop-size distributions, the chemical characteristics of cloud water were investigated. Figure 2 indicates median values of cloud water sulfate obtained during summer from 1986. Three cloud water pH intervals are chosen: pH < 3, $3 < \text{pH} < 3.7$ and

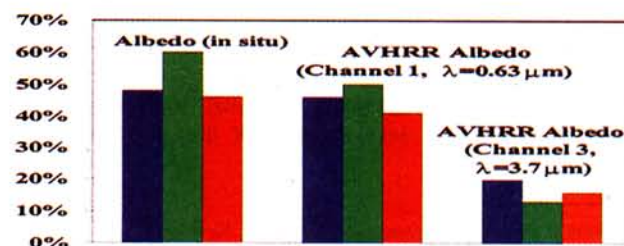


Figure 3b. Average values of cloud albedo calculated from in situ observations and that retrieved from the Advanced Very High Resolution Radiometer (AVHRR) data from Channel 1 at $\lambda = 0.63 \mu\text{m}$ and Channel 3 at $\lambda = 3.7 \mu\text{m}$ for the same three categories as in Fig. 3a.

Table 1. Comparison of differences in the Regression Relations Between: Cloud Condensation Nuclei Concentration (CCN), Cloud Droplet Number Concentration (N), Subcloud Aerosol Number Concentration (N_a) and Sulfates for the Southeastern and Northeastern U.S. NA is for Not Available and cw is for Cloud Water.

Location	N and CCN/ N_a	CCN and sulfate	N and sulfate
Southeast U.S.	$N = 183 \log(\text{CCN}) - 334$	$\text{CCN} = 60.4 (\pm 11) [\text{SO}_4^{2-}] + 29 (\pm 96)$	$\log[N] = 0.66 (\pm 0.05) \log [\text{SO}_4^{2-}]_{\text{cw}} + 0.67 (\pm 0.02)$
Northeast U.S.	NA	NA	$\log[N] = 0.26 (\pm 0.05) \log [\text{SO}_4^{2-}]_{\text{cw}} + 1.95 (\pm 0.21)$
Puerto Rico	NA	NA	$\log[N] = 0.09 (\pm 0.05) \log [\text{SO}_4^{2-}] + 2.32 (\pm 0.15)$
Northeast Atl.	NA	$\text{CCN} = 15.5 (\pm 3.3) [\text{SO}_4^{2-}] + 95 (\pm 16)$	NA
North Atlantic	$N = 216 \log(N_a) - 382$	NA	NA

pH > 3.7 to depict PC, C and M air masses, respectively. The trends in cloud water sulfate depicted in Fig. 2 indicate that sulfates were higher during 1993-97 than during 1986-89. Previous analyses indicated a strong correlation between pH and wind direction of air masses and sulfate with the source of air masses [Deininger and Saxena, 1997; Ulman and Saxena, 1997; Menon et al., 1999]. Acker et al. [1998] also found air masses with lowest acidity and highest sulfate were from polluted areas in Europe at their ground-based observatory in Germany.

Table 1 gives the regression relations between CCN, N, subcloud N_a and sulfates for the southeastern and northeastern U.S. CCN are reported at 1% supersaturations (ss) for comparison with other studies. Usually for clouds encountered at the Mt. Mitchell site we would expect lower ss and correspondingly lower CCN. A quantitative relationship that determines the increase in CCN from an increase in N_a depends on cloud chemical/dynamical characteristics and on aerosol size distribution. Non-linear relationship between CCN-N were obtained from our 1995-96 measurements, similar to non-linear N_a -N relationships given in Gueltepe and Isaac [1996] for the North Atlantic, as in Table 1. Using the relations in Table 1, the change in cloud albedo (ΔA_c) from an enhancement in N or CCN, when N_a increases from 100 to 1000 cm^{-3} (values are chosen to represent clean and polluted air) was calculated using a simple estimate, $\Delta A_c = 0.057 \Delta N/N = 0.04 \Delta \text{CCN}/\text{CCN}$ as in Kaufman and Chou [1993], where ΔN and ΔCCN are increases in N and CCN resulting from an increase in N_a and N. For the southeastern U.S., ΔA_c is 0.33, whereas for the North Atlantic ΔA_c is 0.25. For the southeastern U.S., for the 1995 data set, the range of sulfate mass was 1.5–30 $\mu\text{g m}^{-3}$ and CCN concentrations were 130 to 1700 cm^{-3} , whereas that for the Northeast Atlantic was 0–10.5 $\mu\text{g m}^{-3}$ for sulfates and 50–250 cm^{-3} for CCN [Hegg, 1994]. Using the CCN-sulfate regression in Table 1, ΔCCN was calculated for an increase in sulfate from 2 to 5 $\mu\text{g m}^{-3}$ (representative of average sulfate in clean and polluted air). The calculated ΔA_c is 0.03 for the Southeast and 0.01 for the Northeast Atlantic. The trend in R_{eff} from 1986-97 was calculated for median values of sulfate from Fig. 2 and representative values of N and LWC for PC, C and M air masses. The trend indicates greater differences in R_{eff} between M and PC air masses during 1993-97 (54%) than during 1986-89 (45%). Modeling studies [Jones et al., 1994] find larger cooling takes place for the largest change in R_{eff} implying greater cooling during 1993-97. The apparent sensitivity of change in N to the change in sulfate was estimated from the slope of log N-sulfate regression relation [Novakov et al., 1994]. The higher sensitivity between N-sulfate obtained from our results compared to those at Puerto Rico [Novakov et al., 1994] and in the Northeast [Leitch et al., 1992] (Table 1) suggests that the indirect forcing is higher for the southeastern U.S. Previous analysis [Menon and

Saxena, 1998] of N-sulfate relationship have indicated that the indirect forcing for this region is higher than -4 W m^{-2} that was estimated by Boucher and Lohmann [1995].

The cloud optical depth (τ) is calculated [Twomey, 1977] as

$$\tau = H \{4.5 \pi N (\text{LWC})^2 \rho^{-2}\}^{1/3} \quad (1)$$

where ρ is liquid water density and the cloud thickness, H is obtained from the HY-SPLIT sounding. The cloud albedo (A) is evaluated from Lacis and Hansen [1974] as

$$A = \tau / (\tau + 7.7) \quad (2)$$

A, as evaluated leads to a preference for scattering in the forward hemisphere and is an approximation to reality. However, this would not change the character of the dependence of A on sulfate. A, calculated as in Eq. 2 for thin clouds was compared to that inferred from the Advanced Very High Resolution Radiometer (AVHRR) data as described in Saxena et al. [1996]. During four years (1993-96) a total of 20 cases were obtained for comparison ($r^2 = 0.71$). Figure 3a indicates average values of LWC, N, R_{eff} and cloud liquid water path (LWP) (taken as the product of LWC and H) for three different sulfate contents. Figure 3b depicts average values of A calculated from *in situ* measurements and that from the AVHRR data for Channel 1 at 0.63 μm and for Channel 3 at 3.7 μm . Channel 1 radiance is chiefly composed of reflected solar radiation and depends on N, H and to a smaller extent on LWC. Cloud reflectance at 3.7 μm is a combination of reflected and emitted radiant energy and depends on LWC and R_{eff} [Slingo, 1989]. The sensitivity of reflectance to droplet size is greatest in Channel 3 due to the absorption at this wavelength. The droplet scattering and absorption cross-sections are proportional to its geometric cross-section and volume, respectively. The ratio of scattering to absorption would increase with a decrease in droplet radius. Thus, Channel 3 reflectance would increase due to an increase in backscattering and the change in the scattering to absorption ratio. If LWC decreases, Channel 3 reflectance would increase whereas Channel 1 reflectance would actually decrease [Mineart, 1988]. In Figs. 3a and 3b, for an increase in average sulfate by a factor of 2.5 (for sulfate < 200 to that between 200–700 $\mu\text{eq l}^{-1}$), N and LWC increase (77% and 15%, respectively), but R_{eff} and LWP decrease (7%). The A's calculated from *in situ* measurements and those from Channel 1 data are enhanced (23% and 9%, respectively). The increase in LWC and the small decrease in R_{eff} result in a decrease in Channel 3 reflectance (36%). An increase in sulfate by a factor of 2 (from sulfate < 700 to sulfate < 1200 $\mu\text{eq l}^{-1}$), decreases R_{eff} and LWC (20% and 52%, respectively), and increases Channel 3 reflectance (25%). The sharp decreases in LWP (64%) decrease A, despite an increase in N (34%). Thus, A does not continually increase with increasing sulfate.

Conclusions

Time series observations of sulfate concentrations in cloud water indicate higher concentrations for 1993-97 as compared to that during 1986-89. The calculated change in cloud albedo from differences in CCN and N in cleaner to polluted air masses was higher for the Southeast as compared to the Northeast. Varying levels of sulfate in polluted and marine air masses lead to changes in R_{eff} that are of sufficient magnitude to counteract warming expected due to doubling of CO_2 . Higher sensitivity of N to sulfate is obtained for the southeastern U.S. as compared to that for eastern North America and Puerto Rico. The indirect sulfate forcing for the Southeast is greater than the -4 W m^{-2} estimated by modeling studies [Boucher and Lohmann, 1995]. A linear relationship between cloud albedo and cloud water sulfate was not found (both from satellite retrievals and calculations from *in situ* measurements). Variations in N and R_{eff} with varying sulfate and in dynamical properties such as LWC and H were found to be important in determining variations in cloud albedo. An internal mixture of BC and sulfate reduces the sulfate forcing by 1.1 W m^{-2} . However, despite this reduction, the combination of both the direct (-4.8 W m^{-2}) and indirect ($> -4 \text{ W m}^{-2}$) sulfate forcing for 1993-96 suggest that anthropogenic influences could balance any warming expected from the doubling of CO_2 for the southeastern U.S. On a regional scale, a mild cooling trend is underway for the eastern U.S. [Saxena *et al.*, 1997; Saxena and Yu, 1998]. Since regional climatic changes affect global climatic changes through aerosol-cloud-climate interactions, consequences of the impact of reduced emissions on climate must be carefully investigated before measures are taken to control emissions.

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